Evaluation of pollutant outflow and CO sources during TRACE-P using model-calculated, aircraft-based, and MOPITT-derived CO concentrations.

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Abstract:

Outflow of CO from Asia during March 2001 is evaluated using data from the TRACE-P mission and the MOPITT instrument in conjunction with model-calculated CO from the University of Maryland chemistry and transport model (UMD-CTM). Comparison of model-calculated CO with aircraft measurements indicates that temporal and spatial variations in CO are well captured by the model (mean correlation coefficient of 0.78); however, model-calculated mixing ratios are lower than observed especially below 850 hPa where negative biases of ~60 ppbv were seen. Regression analysis is used to optimize the magnitudes of the bottom-up TRACE-P Asian fossil fuel (ff), biofuel (bf), and biomass burning (bb) CO emission inventories. Resulting Asian correction factors are 1.70 ± 0.36 for ff/bf emissions and 1.00 ± 0.31 for bb emissions. Resulting ff/bf emissions are 29.6 ± 6.2 Tg for March 2001 (323 ± 67 Tg for an entire year). Resulting bb emissions for March 2001 are 18.1 ± 5.4 Tg. Correction factors are lowest (highest) for experiments that assume a high (low) CO yield for the oxidation of anthropogenic and natural hydrocarbons and for experiments that use (do not use) an aerosol-modified OH distribution. Comparison of model-calculated CO with MOPITT measurements supports the results from our regression analysis. Without exception, mean March 2001 model-calculated CO profiles in the TRACE-P region from a simulation with adjusted CO sources are within a standard deviation of mean March 2001 MOPITT-sampled profiles.

1.0. Introduction

The TRACE-P (NASA Transport and Chemical Evolution over the Pacific) aircraft mission was conducted in February-April 2001 to measure chemical outflow from Asia and to relate it quantitatively to its sources [*Jacob et al*., 2003]. Quantifying Asian outflow is essential because eastern Asia is rapidly developing and outflow from Asia impacts atmospheric chemistry, surface air quality, and radiative forcing on intercontinental scales [*Jaffe et al*., 1999; *Jacob et al*., 1999].

Outflow studies focusing on CO have proven to be especially useful. CO is an ideal tracer of continental outflow. Its sources are moderately well known and are highly correlated with the sources of other pollutants, its chemistry is relatively simple, and its lifetime is long enough (a few weeks over summer continental regions; a year at the winter poles [*Holloway et al*., 2000]) to follow a plume as it circles the earth but short enough to ensure large regional differences in concentration. CO simulations by *Allen et al*. [1996b], *Kanakidou et al*. [1999], *Staudt et al*. [2001], *Kiley et al*. [2003], *Lamarque and Hess* [2003], and others have been useful for studying the effects of transport on the redistribution of pollutants.

Carbon monoxide is also of interest in its own right. It plays a major role in the tropospheric O_3 budget. Globally, oxidation of CO by the hydroxyl radical (OH) in the presence of sufficient NO is a significant net source of tropospheric O_3 [*Crutzen*, 1973; *Chameides*, 1978] and a major sink for OH, the atmosphere's main oxidant. Therefore changes in CO concentrations affect O_3 concentrations and the ability of the atmosphere to cleanse itself.

Finally, CO is of interest because relatively plentiful measurements are available. During the TRACE-P period, satellite-based measurements from MOPITT (Measurements of Pollution in the Troposphere) [*Deeter et al*., 2003], aircraft-based measurements from NASA's DC-8 and P-3B, and ground-based measurements from the NOAA CMDL network [*Novelli et al*., 1998] were available.

In this study, CO outflow during March 2001 is studied using data from MOPITT and the two aircraft in conjunction with model-calculated CO from simulations with different resolutions, convective options, and source magnitudes. Our goal is to estimate the corrections to Asian emissions suggested by the combination of model simulations and CO measurements downwind of Asia. We also evaluate the relative contribution of fossil-fuel combustion, biofuel emissions, biomass burning, and hydrocarbon oxidation to the CO budget downwind of Asia.

2. Data and Model Description

2.1. GEOS-3 DAS

The distribution of CO was simulated using the University of Maryland CTM (UMD-CTM), a version of the Goddard CTM [*Allen et al*., 1996a,b] that accepts meteorological fields from uniform- or stretched-grid (SG) versions of the Goddard Earth Observing System Data Assimilation System [GEOS (SG-) DAS]. Meteorological fields from versions 3 of the GEOS DAS [*Hou et al*., 2003] and GEOS SG-DAS [*Fox-Rabinovitz et al*., 2002] drove these calculations.

Parameterizations of penetrative convection, infrared and solar radiation, and the planetary boundary layer used in versions 1-3 of the GEOS-GCM are described in *Schubert et al*. [1993], *Bloom et al.* [1996], *Helfand et al*. [1999], *Hou et al*. [2003] and references therein. The uniform-grid version of the GEOS-3 DAS has a 1º x 1º horizontal resolution and 48 sigma layers in the vertical. Dynamics in the GEOS-3 SG-GCM [*Fox-Rabinovitz et al*., 1997; *Fox-Rabinovitz*, 2000] are calculated on a stretched-grid with a 40-60 km resolution within a region(s) of interest stretching gradually with latitude and longitude outside of this region(s). Diabatic tendencies are calculated on an intermediate 1º x 1º grid and interpolated onto the stretched-grid. Results from simulations of drought (1988) and flood (1993) conditions in the U.S. [*Fox-Rabinovitz*, 2000] indicate that the stretched-grid approach simulates mesoscale features missed by a coarser resolution simulation while maintaining the integrity of larger-scale features. The analysis systems (i.e., The Physical-space Statistical Analysis System (PSAS) [*Cohn et al*., 1998]) of the stretched- and uniform-grid versions of the GEOS-3 DAS are the same (i.e., the analysis in both systems is performed on a uniform 1º x 1º 36-pressure layer grid). This allows the SG-analysis system to use well-tested statistical structures from the uniform grid analysis system. Results from multiple "region-of-interest" simulations with the SG-DAS are described in *Fox-Rabinovitz et al*. [2002].

2.2. UMD-CTM

The current off-line version of the UMD-CTM has 17 sigma layers, 18 pressure layers, a model top of 0.01 hPa, and a sigma-pressure interface of ~242 hPa. Data from the 48-layer GEOS-3 DAS were mapped offline onto this 35-layer grid before use in the CTM. Layers in the troposphere and lower stratosphere match layers in the GEOS DAS. The model top (0.01 hPa) was chosen to match the model top of the GEOS DAS. A sigma-pressure coordinate system was chosen to minimize stratospheric noise while maintaining mesoscale features in the troposphere. Advection is calculated using a generalized (non-uniform grid) version [*Allen et al*., 2000] of *Lin and Rood's* [1996] multidimensional and semi-Lagrangian extension of the piecewise parabolic method (PPM) [*Colella and Woodward*, 1984; *Carpenter et al*., 1990]. Vertical transport of trace gases by deep convection is parameterized using cumulus mass flux and detrainment fields from the GEOS DAS [*Allen et al*., 1996b]. Turbulent mixing is calculated through a fractional mixing scheme [*Allen et al*, 1996a] with $\alpha = 1$, i.e., complete mixing is assumed in the boundary layer. *Park et al.* [2003] describe the complete UMD-CTM modeling system. In this analysis the UMD-CTM was run with parameterized CO chemistry. Chemical production and loss of CO were prescribed in a manner similar to that of *Allen et al.* [1996b] and *Holloway et al.* [2000] (i.e., prescribed OH concentrations [*Spivakovsky et al.*, 2000] were used for computing CO loss and CO production from methane [CH_4]).

The horizontal resolution of the UMD-CTM varies with experiment. These calculations were performed on a uniform 2º (in the north-south) x 2.5º (in the east-west) grid and on a non-uniform grid with a 0.5º x 0.5º resolution in the TRACE-P region (100º-150ºW; 10º-40º N) stretching gradually to 1.88º x 2.19º on the opposite side of the globe. Horizontal fields from the 1º x 1º GEOS-3 DAS were mapped onto the 2º x 2.5º grid before use. No horizontal mapping was performed on the stretched-grid fields. Instantaneous meteorological fields needed for the CO simulations included 00, 06, 12,

and 18 UT surface pressures, temperatures, and *u* and *v* components of the wind. Time-averaged fields included three-hour averaged planetary boundary layer depth, and six-hour averaged cloud mass flux and detrainment. Instantaneous fields were interpolated in time to the transport time. The vertical velocity is calculated kinematically each transport time step (15 minutes) by assuming the vertical velocity is zero at the top of the model and integrating downward.

2.3. Aircraft and MOPITT CO data

Jacob et al. [2003] list the dates and locations of DC-8 and P-3B flights during the TRACE-P mission. The first scientific flight of the DC-8 (flight 4) occurred on February 26 while the last DC-8 flight (flight 20) occurred on April 9. DC-8 flights 7-17 (March 7-31) are considered local. They sampled Asian outflow in the 100°-150° E longitude range. The first scientific flight of the P-3B (flight 4) occurred on February 24 while the last P-3B flight (flight 24) occurred on April 10. P-3B flights 8-19 (March 4-April 2) are considered local. *Jacob et al*. provide a list of the various trace gases that were measured aboard the DC-8 and P-3B. CO measurements (G. Sachse of NASA/Langley was the principal investigator) were taken on both the DC-8 and P-3B. CO was measured with a one second sampling frequency using differential absorption spectrometry [*Sachse et al*., 1987]. Values are believed to be accurate to within a few percent. CO measurements from the TRACE-P DC-8 and P-3B 5-minute merges are used in this study. These data sets are available from NASA Langley Research Center's Distributed Active Archive Center.

The MOPITT instrument aboard the Terra spacecraft measures emitted infrared radiation at 4.7 μm in the atmosphere and reflected infrared radiance in the atmospheric column and permits retrieval of tropospheric CO columns with a vertical resolution of ~3 km and a horizontal resolution of ~22 km. Global coverage is achieved over cloud-free regions every ~3 days with a 10:30 AM local time equator crossing. Retrieved columns represented by values at the surface and on up to 6 fixed pressure levels are available for the TRACE-P period. MOPITT retrievals are calculated using averaging kernels. The kernels describe the sensitivity of the MOPITT instrument to variations in the true column. Validation studies over the tropical ocean indicate that retrieved columns are most sensitive to upper tropospheric ($P < 500$ hPa) and mid-tropospheric ($800 < P < 300$) variations and least sensitive to variations in boundary layer CO [*Deeter et al*., 2003]. The altitude of peak sensitivity varies with surface temperature; CO near the surface becomes more detectable as the surface temperature rises. Therefore daytime retrievals at polluted sites may see more CO than nighttime retrievals. Validation results indicate that the retrievals have an 8-10 ppbv high-bias in the lower troposphere, a 2-5 ppbv high-bias in the mid-troposphere, and a slight negative bias in the upper troposphere and lower stratosphere [*Emmons et al*., 2002].

March 2001 MOPITT data for the TRACE-P region (defined here to be 100°-150°E; 10°-40°N) are used in this study. MOPITT validation studies during the TRACE-P period [*Jacob et al*., 2003] indicate that under clear-sky or scattered-cloud conditions, CO columns from MOPITT agree well with DC-8 sampled columns passed through the MOPITT averaging kernel.

2.4. Model-calculated CO distributions

Several different calculations with the UMD-CTM were performed in order to study the sensitivity of model-calculated CO during the TRACE-P period to variations in fossil fuel / biofuel source (annual or monthly average), biomass burning source (monthly climatology or daily year 2001), OH sink ("standard" or aerosol-modified), resolution (2˚ x 2.5˚ uniform-grid or 0.5˚ x 0.5˚ stretched-grid), and deep convective option (yes or no). The contributions of fossil fuel combustion (ff), biofuel emissions (bf), biomass burning (bb), isoprene and terpene oxidation, and CH_4 oxidation to model-calculated CO were calculated separately in each experiment. Secondary CO emissions due to oxidation of methane, isoprene and terpene were specified as in *Allen et al*. [1996b]. CO production from methanol and acetone oxidation was not included [*Duncan et al*., 2003b].

Table 1 gives details on the various CO simulations. Simulations Base0, BnC0, and Boh0 were initialized July 1, 2000, and run through April 14, 2001. Simulations Bbb0 and SGbb0 were initialized on February 22, 2001, using output from the Base0 simulation and run through April 14. Simulations Ebb0 and Eoh0 were created using output from simulations Bbb0 and Boh0, respectively. In these experiments, additional CO production by the oxidation of anthropogenic volatile organic compounds (AVOCs) is included by multiplying model-calculated CO with a ff, bf, or bb source by 1.20, 1.19, or 1.11, respectively [*Duncan et al*., 2003b]. This approach assumes that CO production by AVOCs is co-located with the location of direct CO emissions.

Emission distributions for ff and bf were taken from global [*Kiley et al*., 2003] and Asian [*Woo et al*., 2003; *Streets et al*., 2003] inventories. Asia was defined as the region extending "from Pakistan in the west to Japan in the east (60º-158º E) and from Indonesia in the south to Mongolia in the North (13º S-54º N) [*Streets et al*., 2003]." *Kiley et al*. [2003] obtained 1º x 1º ff and bf inventories for a TRACE-P CO modeling intercomparison study by superimposing a year 2000 annual average CO emission inventory for Asia [*Streets et al*., 2003] on a global annual average CO emission inventory [*Yevich and Logan*, 2003]. Superimposing *Woo et al.'s* year 2000 monthly average CO emission inventory for Asia on the annual average global inventory created the "Woo/Kiley" inventory. Since ff/bf emissions are largest during the winter, the use of an annual average ff/bf source introduces a low-bias in model-calculated CO entering the TRACE-P period. Annual average (1° x 1°) and March (0.5° x 0.5°) ff/bf emissions are shown in Figures 1a-b. March emissions over Asia exceed annual average emissions by ~6%.

Both year-specific and climatological bb inventories were used in the CO simulations. The "Base" simulations used climatological monthly average global bb emissions [*Duncan et al*., 2003a]. *Heald et al.* [2003a] used this inventory as the basis for a daily inventory for the TRACE-P period. We used a daily bb inventory prepared by *Woo et al*. [2003] for our "non-Base" simulations. *Woo et al.* created their inventory by combining *Streets et al*. [2003] Asian-specific bb inventory with data from the Advanced Very High Resolution Radiometer (AVHRR). Figures 1c-d show year 2001 [*Woo et al*., 2003] and climatological [*Duncan et al*., 2003a] bb inventories for 10°-45° N and 90°-150° E. Clearly, year 2001 emissions over Southeast Asia are lower than normal, while emissions over China are larger than normal. Overall, year 2001 emissions from *Woo et al*. for this latitude/longitude band are ~40% less than the *Duncan et al.* climatology. However, only a portion of this difference (13% according to *Heald et al*. [2003a]) is attributable to interannual variations in biomass burning. The rest is caused by

differences in the *Streets et al*. and *Duncan et al*. bb algorithms. March 2001 Asian bb emissions from the *Woo et al.* inventory are 30% less than same period bb emissions from the *Heald et al*. inventory. Figure 2 compares daily variations in the *Woo et al*. bb source to monthly average variations in each source. Considerable day-to-day variability is evident. Emissions peak on March 7, a period when burning in Laos (100º E, 19º N) is especially large. Emissions decrease towards the end of TRACE-P period.

Reaction with OH is the primary loss mechanism for CO. OH concentrations are sensitive to aerosol loading. The effect of N_2O_5 hydrolysis in aerosols on OH concentrations is well known and included in the *Spivakovsky et al*. [2000] OH distribution. However, scattering and absorption of ultraviolet radiation and reactive uptake of HO_2 by aerosols also affect OH concentrations. These processes reduce the OH source by decreasing the O_3 photolysis rate and increase the OH (technically the HO_x) sink via uptake of HO_2 by aerosols. The importance of these additional aerosol effects was considered in simulations Boh and Eoh that used modified OH distributions. In these "additional aerosol" simulations, monthly average OH concentrations from *Spivakovsky et al.* were adjusted at each grid point by a factor equal to the ratio between monthly average OH in a GEOS-CHEM [*Bey et al*., 2001] calculation [*Martin et al*., 2003] that included only N_2O_5 hydrolysis in aerosols to monthly average OH in a GEOS-CHEM calculation that also included scattering and absorption of ultraviolet radiation by aerosols and reactive uptake of HO_2, NO_2, and NO_3 by aerosols. Mathematically,

$$\text{OH (mod)} = \text{OH (Spiv.)} \; * \; \frac{\text{OH (GEOS-CHEM [enhanced aerosol])}}{\text{OH (GEOS-CHEM [standard aerosol])}} \qquad (1)$$

Martin et al. [2003] calculated the effects of these processes on OH and CO concentrations. They found that boundary layer OH concentrations are reduced by 5-35% over most of the Northern Hemisphere and by up to a "factor of 4 over India during March." Mixing ratios of CO increased by 5-15 ppbv in most of the Northern Hemisphere. *Tang et al.* [2002] used a regional CTM to simulate the effect of biomass burning on OH concentrations downwind of regions of burning. They found OH decreases of up to 40% in the lowest 3 km of the atmosphere.

Results from experiment SG0 were submitted to the TRACE-P CO Intercomparison [*Kiley et al*., 2003] and are not discussed here. When model to measurement comparisons were performed for each of five layers, the UMD-CTM showed the lowest root-mean-square (RMS) error among all the models in all layers except below 850 hPa. Correlations between model-calculated and measured (DC-8 Florida State University merge) CO were typically between 0.6 and 0.8 but were as low as 0.15 during DC-8 Flight 10.

3. Comparison of Model-calculated and observed CO

CO outflow from Asia during the TRACE-P period consisted of a mixture of anthropogenic and natural sources [*Liu et al*., 2003]. Model-calculated (experiment Bbb0; year 2001 bb source) and DC-8 measured CO outflow on March 10, 2001, (DC-8 flight 9) is shown in Figure 3. Oxidation of hydrocarbons (Figure 3c) contributed a 30-60 ppbv CO background along the flight track. Low-level ff/bf CO plumes (Figure 3a) and

mid-level bb CO plumes (Figure 3b) are evident in the East China Sea and Yellow Sea outflow. The DC-8 sampled the top of the ff/bf plume at 3:30, 4:20, and 5:15 UT and caught the edge of the bb plume at 5:45 and 7:30 UT. The clear separation of the ff/bf and bb CO sources suggests that it may be possible to use CO simulations to infer the relative magnitudes of ff/bf and bb sources.

CO from ff/bf and bb emissions were also clearly separated during several other flights including DC-8 Flight 10 (Figure 4), which sampled Asian outflow into the East China Sea (18º-26º N) [see *Jacob et al.* [2003] for flight track]. During this flight, oxidation of hydrocarbons contributed a background of 40-50 ppbv of CO. Contributions from ff/bf and bb varied greatly with time (location). The DC-8 aircraft encountered ff/bf and bb outflow as it performed two spirals between hours 4 and 6. Observations indicated that the magnitude of low-level outflow exceeded the magnitude of mid-level outflow; CO mixing ratios decreased as the plane ascended and increased as the plane descended. However, model performance during this flight varied significantly with simulation. In simulation Base0 (*a priori* sources; climatological bb), model-calculated mixing ratios were too low in the ff/bf plume and too high in the bb plume (Figure 4a). The correlation ($r = 0.38$) between model output and aircraft measurements was low because in contrast with the measurements model-calculated mixing ratios increased as the plane ascended and decreased as the plane descended. In simulation Bbb0 (*a priori* sources; year 2001 bb), model-calculated mixing ratios were too low in the ff/bf plume but reasonable in the bb plume (Figure 4b). Correlations increased to 0.76. In simulation Bbb2 (*a posteriori* sources calculated as shown in section 3.2; year 2001 bb), observed CO variations were well captured within both plumes. The mean negative bias between model-calculated and aircraft-observed CO decreased from 26.8 to 1.5 ppbv.

Optimization of CO source magnitudes also revealed that errors in the vertical distribution of CO sources must be minimized before assessing the quality of deep convective mixing algorithms. Figure 4d shows the results from simulation BnC0 (*a priori* sources; year 2001 bb; no deep convection). As expected, model-calculated mixing ratios in the upper troposphere are much lower than observed; the mean bias for all data points was 36.7 ppbv. However, the correlation between model output and data for this flight is 0.81! Correlations are high because mixing ratios in this run consistently increase (decrease) with increasing (decreasing) pressure as observed. The high correlations for this flight show that other statistics in addition to correlations must be used when evaluating model performance. They also show that biases in CO sources minimize the positive impact of changes in model transport and/or convective mixing algorithms.

3.1. Evaluation of model performance

Clearly for DC-8 flight 10, increasing the ff/bf source and using year 2001 bb emissions improves the agreement between model-calculated and observed CO. Model performance for various experiments will be evaluated by comparing biases, centered RMS differences (the RMS difference after removing the bias) [RMS_c], standard deviations (σ), and correlations (r) as a function of layer and flight. Figure 5 shows mean biases between model-calculated and aircraft-observed CO for five different vertical layers before and after adjusting CO sources. Before optimization each of the model runs has a substantial negative bias in the lower- and middle-troposphere. Negative biases

continue into the upper troposphere in all but experiments Base (climatological bb source) and Eoh (year 2001 bb source; AVOC source; aerosol modified OH). The inclusion of an AVOC CO source reduces biases by ~25 ppbv in the PBL and by 5-10 ppbv in the upper troposphere. The inclusion of additional aerosol effects reduces CO biases along the DC-8/P-3B flight tracks by ~8 ppbv in the boundary layer and by ~5 ppbv in the upper troposphere. Changes to r and RMS_c (not shown) are small as expected given the lifetime of CO

In Figure 6, "Taylor" diagrams [*Taylor*, 2001] are used to compare RMS_c, σ, and r for the same layers from simulations Bbb0 [year 2001 sources] (a), Base0 [climatological sources] (b), SGbb0 [0.5° x 0.5°; year 2001 sources] (c), and BnC0 [no deep convection; year 2001 sources] (d). In most layers, correlations increase and RMS_c decrease with increasing horizontal resolution and with the use of year 2001 sources. Increasing the horizontal resolution is most beneficial below 850 hPa and for $500 < P < 300$ hPa. For example, below 850 hPa, r increases from 0.70 to 0.80 and RMS_c decreases from 73 to 67 ppbv. Increasing the horizontal resolution degrades model performance in layer 2 ($850 < P < 700$ hPa). In this layer, r decreases from 0.67 to 0.62 and RMS_c increases from 59 to 62 ppbv. Introduction of year 2001 sources has a beneficial impact in all layers except layer 4 ($500 < P < 300$ hPa). For example, for $P < 300$ hPa, r increases from 0.45 to 0.55, and RMS_c decreases from 34 to 30 ppbv.

Figures 5 and 6c-d show the impact of deep convection on model performance. As expected, excluding deep convection increases concentrations in the boundary layer and decreases concentrations in the middle- and upper-troposphere. Low-biases are reduced by 3-5 ppbv in the boundary layer but are increased by 25-30 ppbv in the middle- and upper-troposphere. Variability in the uppermost troposphere ($P < 300$ hPa) is reduced by a factor of three, while variability in the boundary layer is unchanged. Variability in layer 3 ($700 < P < 500$ hPa) increases by more than 50%.

3.2. Estimation of biases in ff/bf and bb CO sources

Qualitative information on trace gas sources can be obtained by comparing model-calculated trace gas concentrations with measurements [e.g., *Pinto et al.*, 1983; *Holloway et al*., 2000]. More quantitative information on the spatial and temporal distributions of sources can be obtained through formal three-dimensional inversion studies [e.g., *Bergamaschi et al*., 2000a, b; *Kasibhatla et al*., 2002; *Petrón et al*., 2002; *Palmer et al*., 2003; and *Arellano et al*., 2003].

In this study, regression analysis is used to estimate the total magnitude of ff/bf and bb CO emissions. Model-calculated CO from simulations with *a priori* sources are used in conjunction with data from local aircraft flights and the MOPITT instrument. Model-calculated CO (**Model**) at any location and time (**i**) contains contributions from **ff**, **bf**, **bb**, and hydrocarbon oxidation (**hc**). Symbolically,

$$\mathbf{Model_i = ff_i + bf_i + bb_i + hc_i,} \qquad \mathbf{(2a)}$$

where $\mathbf{hc}_i$ in this study identifies CO from the oxidation of methane, isoprene, and terpene. These contributions may also be subdivided into Asian and non-Asian terms. Symbolically,

$$\mathbf{Model_i = ffas_i + ffrow_i + bfas_i + bfrow_i + bbas_i + bbrow_i + hc_i,} \quad \mathbf{(2b)}$$

where the ff, bf, and bb terms are broken into Asian (**as**) and rest-of-world (**row**) components.

In order to estimate global corrections to the CO sources suggested by the aircraft- and satellite-data, the regression model

$$\mathbf{y_i \equiv Data_i - Chc_i = A1(ff_i+bf_i) + B1(bb_i) + \varepsilon 1_i,} \quad \mathbf{(3a)}$$

was fitted to the model-calculated and observed CO mixing ratios. In order to estimate the Asian corrections to CO sources suggested by the data, the regression model

$$\mathbf{y_i \equiv Data_i - Chc_i - ffrow_i - bfrow_i - bbrow_i = A2(ffas_i+bfas_i) + B2(bbas_i) + \varepsilon 2_i,} \quad \mathbf{(3b)}$$

was fitted to the model-calculated and observed CO mixing ratios. The observed response [y_i] (the left hand side of equations 3a or 3b) is equal to the fitted response [e.g., $\mathbf{A1(ff_i+bf_i) + B1(bb_i)}$] plus an error term ($\boldsymbol{\varepsilon}$). Coefficients A1, B1, A2, and B2 are scaling factors that when applied to the appropriate combination of CO source types minimize the sum of squares of the deviations of the observed responses from the fitted responses. CO with a hydrocarbon source ($\mathbf{hc_i}$) multiplied by a constant **C** appears on the left hand side of equations 3a and 3b. This hydrocarbon source is not optimized. The sensitivity of the ff/bf and bb source optimizations to uncertainties in the source of CO via oxidation of natural hydrocarbons (CO source via AVOC oxidation is not multiplied by **C**) is evaluated by fitting equations 3a and 3b for $\mathbf{hc_i}$ equal to 80% (**C** = 0.8), 100% (**C** = 1.0), and 120% (**C** = 1.2) of its model-calculated value. Fossil fuel and biofuel CO are grouped together because of their similar spatial distributions. Biomass burning CO is treated separately.

3.2.1. Fit to aircraft data

Equations 3a-b were applied to model-calculated CO and data along local DC-8/P-3B flight tracks. Table 2 shows correction factors suggested by the fits to model-calculated and aircraft observed data and the resulting goodness of fits (i.e., the fraction of the variance in the observed data set explained by model-calculated CO before and after optimization). Before optimization, simulation Eoh (AVOC emissions; modified OH) provided the best fit and simulation Base (climatological bb source) provided the worst fit. After optimization, simulation SGbb (0.5º x 0.5º) provided the best fit and simulation BnC (no deep convection) provided the worst fit. The remaining simulations had similar goodness of fits. Comparison of pre- and post-optimization goodness-of-fits indicates that optimization compensates for biases in the magnitude of CO sinks and sources. These source/sink biases mask errors in model-calculated vertical mixing and limit the gain possible when the resolution is increased. Or to put it another way, increasing horizontal resolution or neglecting deep convective mixing is most beneficial/detrimental after optimization of source terms.

The global ff/bf correction factors range from 1.20 to 1.73. These values imply that a better fit to the TRACE-P aircraft data could be obtained if the global ff/bf source in the UMD-CTM was increased by 20-73%. Since most ff/bf outflow occurs in the

boundary layer, the magnitude of the necessary ff/bf correction is proportional to the magnitude of the bias between model-calculated and observed CO in the surface-to-850 hPa layer (see Figure 5). Therefore correction factors are smaller for calculations with larger CO sources and smaller CO sinks. For example, the Eoh correction is smallest because this simulation includes CO from oxidation of AVOCs and uses an aerosol-enhanced OH distribution. Corrections to bb emissions vary with emission source and OH distribution. As anticipated, corrections are largest when climatological emissions (experiment Base) are used. In these simulations, best agreement is obtained when global (Asian) bb emissions are reduced to 69 ± 10% (55 ± 10%) of their climatological value. Relatively modest corrections (1.19 ± 0.19 for experiment Bbb) to bb emissions are required for simulations using year 2001 sources. Corrections are even smaller (1.03 ± 0.19 for experiment Boh) when additional aerosol effects are included. Including these effects reduces CO loss via reaction with OH; Biases are reduced and corrections reduced.

Heald et al. [2003b] demonstrate that biases between model-calculated and MOPITT-observed Southeast Asia CO profiles can be caused by either biases in model-calculated deep convective mixing or biases in Southeast Asia CO sources. Required correction factors are also impacted by these biases. Model-calculated mean latitudinal and vertical CO profiles from experiments Bbb (2.0º x 2.5º) and SGbb (0.5º x 0.5º) are compared to aircraft measurement derived profiles in Figures 7a-d. As expected, given the *a priori* underestimation of the ff/bf sources, model-calculated CO in both *a priori* simulations (the dotted lines) is considerably lower than observed especially at pressures of greater than 800 hPa and at latitudes northward of 26° N. Boundary layer mixing ratios in the 0.5º x 0.5º simulation are ~7 ppbv less than boundary layer mixing ratios in the coarser resolution simulation, while 500-350 hPa mixing ratios in the stretched-grid simulation are ~7 ppbv more than 500-350 hPa mixing ratios in the 2.0º x 2.5º run. Clearly CO from simulation SGbb0 falls off less rapidly with height than CO from experiment Bbb0. These differences indicate that vertical mixing in the 0.5º x 0.5º simulation exceeds vertical mixing in the 2.0º x 2.5º simulation. Since deep convection is the primary mechanism that transports Asian biomass burning emissions to the Pacific [*Liu et al*., 2003], the *a posteriori* bb source in experiment SGbb2 (B = 0.77 ± 17) is less than the *a posteriori* bb source in experiment Bbb2 (B =1.19 ± 0.19).

3.2.2. Fit to MOPITT data

The MOPITT instrument also measured CO during the TRACE-P period. In order to evaluate the robustness of our conclusions in section 3.2.1, we re-calculated Asian corrections with equations 3a-b using MOPITT data and model output from simulation Bbb. Global corrections to the CO sources suggested by MOPITT data were calculated as follows: For the time period, March 1-31, 2001, 107,166 CO profiles were taken by MOPITT in the region defined by 10º-40ºN and 110º-150ºE. Interpolating model output in the horizontal grid boxes containing these MOPITT observations to the pressures and times of the observations, model data sets were created on the MOPITT vertical grid (the surface, 850, 700, 500, 350, 250, and 150 hPa) for total CO and for CO from each source type. The MOPITT averaging kernel was then applied to the model-calculated profiles. Since the *a priori* profiles used in the MOPITT retrievals are for total CO, it was necessary to multiply model-calculated source type profiles by a constant

(the ratio of total model-calculated CO at 700 hPa to CO from a particular source type) before applying the kernel. Symbolically,

$$cX'_{comp} = X_a + A(cX_{comp}-X_a), \quad (4)$$

where X'_{comp} is the resulting transformed model-calculated CO profile, X_{comp} is the "untransformed" model-calculated CO profile, X_a is the *a priori* MOPITT CO profile, A is the averaging kernel, which is calculated using the *a priori* covariance matrix and the retrieved CO error covariance matrix, and c is a constant that varies with source type and equals 1 when total CO is transformed. Version 3 *a priori* CO profiles and covariance matrices were taken from the MOPITT website (www.eos.ucar.edu/mopitt). The retrieved CO error covariance matrix is specific to each MOPITT observation and is read in with the MOPITT data. The resulting profiles are weighted averages of the model-calculated profile and the *a priori* profile. Differences between the transformed total CO profile and the sum of transformed CO source type profiles did not exceed 3%. Since the resulting data sets are highly correlated in the vertical (due to the averaging kernel), it is not desirable to use output from all seven layers in the regression analysis. In order to sample copious ff/bf and bb outflow, we use 700 hPa MOPITT data for $\mathbf{Data_i}$ and 700 hPa transformed model output for each source type $\mathbf{ff_i}$, $\mathbf{bf_i}$, etc. in equations 3a-b.

Asian correction factors calculated using MOPITT observations were 2.00 ± 0.26 for ff/bf emissions and 0.83 ± 0.17 for bb emissions. Applying these correction factors to the 700 hPa fields, reduced the negative bias of the model from 21.5 to 4.8 ppbv, increased 1-SSE/SST from 0.946 to 0.967, and increased model-calculated standard deviations from 19.8 to 28.1 ppbv. Model performance was not improved uniformly. Applying the correction factors decreased the correlation coefficient slightly (0.68 to 0.67) and increased RMS_c slightly (22.6 to 24.0 ppbv). Applying the aircraft-based correction factors to the 700 hPa MOPITT data/output, resulted in similar changes (a negative bias of 2.2 ppbv, a σ of 28.5 ppbv, and a 1-SSE/SST of 0.966).

The MOPITT- and aircraft-based (1.96 ± 0.10) ff/bf corrections differ only slightly. Therefore this regression analysis using MOPITT data provides additional evidence that the *a priori* ff/bf source is too low. The MOPITT- and aircraft-based (1.10 ± 0.21) bb corrections differ considerably. Comparison of 700 hPa model output with MOPITT data suggests that the year 2001 bb source is one-sixth too low. Comparison of model output with aircraft data suggests that the year 2001 bb source is 10% too high. Several different explanations are possible for the different estimates. MOPITT and the aircraft sample different locations at different times. In particular, MOPITT only samples partly cloudy and/or cloud-free time periods. Periods during which frontal lifting bring bb outflow to the Pacific may be underrepresented. This sampling bias could cause MOPITT-based estimates of bb-CO to have a low-bias (see section 3.4) [*Crawford et al.*, 2003] and lead to an underestimate of the bb correction factor.

3.3. Evaluation of CO outflow with aircraft observations

Vertical and latitudinal biases in the *a posteriori* simulations are much smaller than in the *a priori* simulations (Figure 7). The much lower biases are expected since the regional (global) magnitude of the ff/bf and bb sources in experiments Bbb (SGbb) were tuned using the aircraft data! However, the extent of the impact is encouraging and

suggests that the spatial distribution of the *a priori* CO sources is better known than the absolute magnitude of emissions. The contribution of individual source terms (ff, bf, and bb) is also shown for the *a posteriori* simulations. Contributions from ff and bf increase with latitude and decrease with height, although the slopes for ff CO greatly exceed the slopes for bf CO. Biomass burning CO is most important in the mid- and upper-troposphere between 15° and 30° N. The use of *a posteriori* sources reduced absolute biases to less than 5 ppbv throughout the troposphere. A slight positive bias was introduced in the upper troposphere (P < 300 hPa) [see Figure 5].

Figure 8 summarizes the contribution of various CO source types (8a) and source types/regions (8b) to model-calculated CO (experiment Bbbr2; *a posteriori* Asian sources) along all P-3B flight tracks. Overall, ff combustion is the most important source of model-calculated CO. Contributions from bb were most significant during Flights 9, 10, and 13. Biofuel- and ff-CO are highly correlated from flight to flight (r=0.92). The ratio of bf/ff CO ranges from 0.25 for flight 17 to 0.42 for flight 7. Flight-to-flight variations in the contribution of methane and NMHC oxidation are minor. Averaging over local P-3B flights, Asian sources account for ~69% of ff/bf CO and ~65% of bb CO. Asian ff/bf sources are most (least) important during flight 9 (flight 17) when they account for 77% (59%) of total ff/bf CO. Asian bb sources are most (least) important during flight 10 (flight 15) when they account for 81% (40%) of total bb CO.

Taylor diagrams comparing model-calculated and observed CO time series along DC-8 flight tracks 7-17 and P-3B flight tracks 8-19 are shown in Figures 9a-b and 9c-d, respectively. In general, optimization led to marginally improved correlations and much more realistic standard deviations (σ) and RMS_c. Optimization increased the mean σ for the 23 local DC-8/P-3B flights to 64 ppbv from 48 ppbv. The mean measured σ was 76 ppbv. Variability was most underestimated during DC-8 flight 13 and P-3B flight 14. During DC-8 flight 13, measured CO mixing ratios in low-level outflow to the East China Sea (downwind of Shanghai) approached 1000 ppbv. During P-3B flight 14, measured CO mixing ratios in low-level outflow to the Yellow Sea exceeded 800 ppbv. The model greatly underestimated the magnitude of these plumes. Indeed these plumes are very difficult to simulate with coarse resolution models. *Kiley et al*. [2003] found that all models participating in the TRACE-P CO intercomparison underestimated the magnitude of the largest outflow events by at least a factor of two. DC-8 flight 14 was the worst simulated of the local flights. The goal of this flight was to sample convective outflow. CO mixing ratios of 200-250 ppbv were observed in high-level outflow to the northwest of Guam. The model simulations did not capture this outflow.

Figures 10a-b evaluate model performance for the DC-8 flights as a function of resolution. Overall, increased resolution resulted in higher correlations (0.78 vs 0.84), lower RMS_c (48 vs 45 ppbv), and slightly lower σ (61 vs 60 ppbv). The largest beneficial impact was seen during DC-8 flight 13; a low-level ff plume and a mid-level mostly bb plume were captured more accurately by the higher resolution simulation. *A posteriori* correlations were 0.92 vs 0.81 for the lower resolution simulation. RMS_c decreased from 91 to 70 ppbv. Increased resolution led to poorer model performance during DC-8 flights 10 and 14. The mixed performance was not surprising. Correlation coefficients for mesoscale models participating in the TRACE-P CO intercomparison were often less than those of coarser resolution global models [*Kiley et al*., 2003].

3.4. Evaluation of CO outflow with MOPITT data

Figures 11a-d show four representations of the mean 2° x 2.5° CO column during March 2001. The mean March 2001 columns were obtained by averaging MOPITT observations and model-calculated output within each model grid box. Comparison of Figures 11a and b provides additional evidence that model-calculated ff/bf sources are underestimated; Model-calculated CO columns poleward of 30º N are considerably less than observed. Much better agreement is obtained when *a posteriori* aircraft-comparison based sources from experiment Bbbr2 are used (Figure 11c). Mean model-calculated CO before passing through the averaging kernel is shown in Figure 11d. Horizontal gradients are sharper and local magnitudes greater. One can see very large column amounts over China. Here the large contribution from the lower troposphere is not well detected by MOPITT.

Figures 12a-o compare mean March 2001 model-calculated CO profiles from simulations Bbb0 and Bbbr2 with MOPITT profiles. Mean model-calculated profiles from simulation Bbbr2 remain within a standard deviation of measurements throughout the TRACE-P region (100°-150° E). Differences are largest over southwestern China (105º E, 25º N) where the model-calculated mean profile is almost a standard deviation larger than the MOPITT-sampled mean profile. With the exception of southwestern China, mean model-calculated profiles from simulation Bbb0 are too low. Differences exceed one standard deviation for the 30º-40º N, 120º-150º E region.

Aircraft-sampled CO mixing ratios during TRACE-P varied with cloud cover [*Crawford et al*., 2002]. Under cloudy conditions CO amounts in the 1-5 (5-11) km layer exceeded clear-condition amounts by 32 (15%). MOPITT only obtains samples during cloud-free or partly cloudy periods. Therefore sampling biases exist because cloud-free regions are sampled more frequently than cloudy regions. *Crawford et al*. [2002] estimated that clouds covered 50-70% of the TRACE-P region. These values imply that MOPITT underestimates 1-5 (5-11) km outflow by 15-22 (7-10)%. However, a comparison of the average GEOS-CHEM CO column during TRACE-P with the average column during MOPITT sampling periods did not reveal a bias [*Heald et al*., 2003b].

The effect of sampling biases on model-calculated CO with an Asian ff/bf or Asian bb source can be seen by comparing the differences between the dashed (sampling at location of MOPITT profiles) and dotted (sampling at all grid points in the latitude/longitude range) lines in Figures 13 and 14. Differences are largest in the lower troposphere, especially during periods of strong low-level outflow. In the lower troposphere, at most locations, sampling during periods when MOPITT was observing resulted in an underestimation of ff/bf-CO (dashed line is to left of dotted line). In the upper troposphere, differences for ff/bf-CO were small. Upper tropospheric differences for bb-CO vary with latitude. At 15º N, sampling biases cause MOPITT to underestimate the contribution of bb-CO to CO outflow (i.e., model-calculated upper tropospheric CO with an Asian bb source was larger during cloudy periods than during partly cloudy and/or clear periods). Differences approached 50% in some longitude bins. At 35º N and at oceanic grid points, sampling biases lead to a 20-30% overestimation of CO. At these locations, CO with an Asian bb source was smaller during cloudy periods than during other periods. Absolute differences are fairly small as most bb outflow occurs at more southerly latitudes.

The effect of MOPITT processing on a CO column varies with the vertical distribution of tracer. Since the MOPITT weighting function peaks in the mid- and upper-troposphere, deviations from X_a, the *a priori* MOPITT CO profile, are underestimated in the lower troposphere and overestimated in the mid- and upper-troposphere. The effect of MOPITT-processing on model-calculated CO with a ff/bf or bb source can be seen by comparing the difference between the dashed (unprocessed) and solid (processed) lines in Figures 13 and 14, respectively. Strong low-level ff/bf outflow (model-calculated Asian ff/bf-CO exceeding 200 ppbv) is evident in several of the regions. MOPITT misses this outflow; Processed low-level CO mixing ratios are much less than their unprocessed counterparts. Differences between MOPITT-processed and unprocessed CO profiles are much smaller during periods of limited low-level outflow. During these periods, actual outflow does not differ greatly from X_a in the lower troposphere. At oceanic longitudes, Asian bb-CO outflow often peaks above the boundary layer (e.g., Figure 14; 135º E, 15º N). MOPITT with its limited vertical resolution and lower tropospheric sensitivity has no hope of capturing this feature.

4.0. Discussion

Table 2 provides several different estimates for the global and regional ff/bf and bb correction factors needed to minimize the differences between model-calculated and measured CO mixing ratios along DC-8/P-3B flight tracks. From Table 2 we see that Asian ff/bf correction factors for simulations with monthly average ff/bf and daily average bb emissions (the non-Base simulations) range from 2.06 for simulation Bbbr (Spivakovsky OH; no AVOC's) when a hydrocarbon oxidation constant (**C**) of 0.8 is assumed to 1.42 for simulation Ebbr (Spivakovsky OH; AVOC's) when **C** is assumed to be 1.2. Additional aerosol effects were not considered in the regional simulations. However, they do reduce the global correction factor by a factor of 1.20 / 1.26 (A[Eoh] / A[Ebb]). Applying this factor as a regional correction gives us a lower bound of 1.35 for the Asian ff/bf correction. Using these bounds, our best estimate for the Asian ff/bf correction is 1.70 ± 0.36. When this correction factor is applied to the *a priori* March ff/bf source (17.4 Tg) we obtain an *a posteriori* Asian ff/bf source strength of 29.6 ± 6.2 Tg. Applying the March correction factor to annual ff/bf emissions yields an *a posteriori* Asian annual source of 323 ± 67 Tg (The *a priori* annual source is 189 Tg). Assuming 48% of Asian emissions originate in China (table 1 of *Palmer et al*. [2003]) we obtain 155 ± 32 Tg as our estimate for annual Chinese ff/bf emissions.

Asian bb correction factors for calculations with a year 2001 bb source range from 0.80 to 1.31. Including additional aerosol effects reduces the required global correction by a factor of 0.93 / 1.07. Multiplying this factor by 0.80 gives us a lower bound of 0.69 for the Asian bb correction. Therefore our best estimate for the Asian bb correction is 1.00 ± 0.31. When this correction factor is applied to the *a priori* March 2001 bb source (18.1 Tg) we obtain an *a posteriori* March 2001 bb source of 18.1 ± 5.4 Tg.

Figures 15a-b compare our estimates for Chinese ff/bf and Asian bb emissions to other recent estimates. *Streets et al*. [2003] using a bottom up approach obtained a year 2000 ff/bf source of 109 ± 61 Tg. Their estimate is lower than other recent estimates. They note that uncertainties in coal use may contribute to a low-bias in their estimate.

Their year 2000 values were calculated using official energy statistics that show large decreases in household coal use between 1995 and 2000. While these decreases were observed in parts of China, actual decreases in central China are now believed to be occurring at a slower rate. *Carmichael et al*. [2002] determined that increasing domestic-sector CO emissions in central China in the Streets et al. inventory by a factor of 2-5 minimizes differences between observed and model calculated CO mixing ratios. Applying this factor to their Chinese source yields a source of 198 ± 30 Tg [*Palmer et al*., 2003]. *Palmer et al*. [2003] in an inverse modeling study using TRACE-P aircraft observations and model-calculated CO from the GEOS-CHEM estimated the Chinese source to be 168 ± 5 Tg. *Petron et al*. [2002], in an inverse modeling study for 1990-1996 estimated the Chinese source to be 262 ± 69 Tg CO yr^{-1} (obtained from their Table 2 by multiplying the sum of the Asian technological and agricultural waste burning and fuelwood use source by 0.48). *Kasibhatla et al*. [2002], in a study using GEOS-CHEM model output and CO data from the NOAA/CMDL Network estimated the ff/bf source in China contributed 175 ±7 Tg of CO in 1994 (obtained by multiplying their estimate for Asia by 0.48). *Palmer et al*. [2003] give possible explanations for discrepancies between these other recent estimates.

Our model analysis does not support any changes to Woo et al.'s estimate (18.1 Tg ± 50%) for March 2001 Asian bb emissions. *Heald et al.* [2003b] used a larger March 2001 bb source (25.4 Tg) in a CO simulation with GEOS-CHEM. They found that model-calculated CO columns over Southeast Asia were overestimated by 18% relative to MOPITT indicating that Southeast Asian bb emissions needed to be reduced by 50-60%. This reduction if applied over all of Asia would imply a biomass-burning source of 10-15 Tg. However, year 2001 emissions over China are greater than climatology (see Figure 1) indicating that their actual value for all of Asia is fairly close to our estimate. *Palmer et al*. [2003] in an inverse modeling study estimated that *Heald et al.*'s year 2001 Southeast Asia and India bb emissions are too large by approximately a factor of three. They obtained an Asian bb source of approximately 8 ± 2 Tg.

5. Summary

Outflow of CO from Asia during March 2001 was evaluated using data from the TRACE-P mission and the MOPITT instrument in conjunction with model-calculated CO from the University of Maryland CTM. Comparison of model-calculated CO with aircraft measurements was encouraging. Temporal and spatial variations in CO were fairly well captured (mean correlation coefficient of 0.78); however, model-calculated mixing ratios were lower than observed especially below 850 hPa where negative biases of ~60 ppbv were seen. Additional aerosol effects explain a portion of the negative biases. Below 300 hPa, biases between model-calculated and observed CO distributions were reduced by 6-9 ppbv when OH concentrations were adjusted to take into the account the effect of aerosols on photolysis- and uptake-rates.

The use of year 2001 bb emissions was critical. Biomass burning emissions during March 2001 were less than climatological emissions. Increasing the resolution from 2.0° x 2.5° to 0.5° x 0.5° resulted in a slightly better overall simulation; Improvement was largest in the boundary layer. Model-calculated upper tropospheric

CO concentrations in the absence of deep convection were 25-35% lower than concentrations calculated with deep convection.

Regression analysis was used to estimate the corrections to Asian CO emissions suggested by the combination of model simulations and aircraft measurements in the TRACE-P region. Resulting Asian correction factors were 1.70 ± 0.36 for ff/bf emissions and 1.00 ± 0.31 for bb emissions. Resulting ff/bf emissions were 29.6 ± 6.2 Tg for March 2001 (323 ± 67 Tg for an entire year). Resulting bb emissions for March 2001 were 18.1 ± 5.4 Tg. The lower emission bounds were found for calculations that included AVOCs, an aerosol-modified OH distribution, and assumed a hydrocarbon oxidation correction factor of 1.2. The upper emission bounds were found for calculations without AVOCs, that included standard Spivakovsky OH, and assumed a hydrocarbon oxidation correction factor of 0.8. These simulations support the results of recent studies indicating that Asian sources of ff and bf are underestimated in bottom-up inventories. These simulations do not support any changes to bottom-up estimates of bb emissions.

Comparison of model-calculated CO with MOPITT measurements supported the results from our regression analysis. Without exception, mean March 2001 model-calculated CO profiles in the TRACE-P region from a simulation with adjusted CO sources were within a standard deviation of mean March 2001 MOPITT-sampled profiles. Biases introduced by applying the MOPITT kernel and sampling during cloud-free periods and/or partly cloudy periods were examined. At 15º N, sampling biases caused MOPITT to underestimate by 30-50% outflow of Asian CO with a bb source.

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Figures

Figure 1. Direct CO emissions: a) 0.5º x 0.5º ff/bf source for March 2000, b) 2.0º x 2.5º annually averaged climatological ff/bf source, c) 1.0º x 1.0º bb source for March 2001, d) 2.0º x 2.5º climatological bb source for March. Units are molecules $cm^{-2}s^{-1}$ (scaled by 1.e-11).

Figure 2. Biomass burning emissions (Tg CO $month^{-1}$) as a function of time (February 22- May 7) for the 90º-150ºE; 10º-45ºN. Solid line shows year 2001 emissions (Woo et al.) Dotted line shows monthly mean of this source. Dashed line shows climatological monthly average bb source of Duncan et al.

Figure 3. Model-calculated contributions (experiment Bbb0) of ff/bf emissions (a), bb emissions (b), and total hydrocarbon emissions (c) to total CO (d) for DC-8 Flight 9. Asterisks show aircraft-sampled CO with the size of the asterisks proportional to the magnitude of observed CO. The longitudes (latitudes) covered by the measurements are shown with a solid (dashed) line.

Figure 4. Model-calculated versus measured CO for DC-8 Flight 10 (March 13, 2001). Asterisks show CO data from the 5-minute merge. Shading shows model-calculated contribution from each source term. Solid line shows sampling pressures. Numbers following the title of each source term give the percent contribution of each term to total CO along the flight track. Results from experiments Base0 (a), Bbb0 (b), Bbb2 (c), and BnC0 (d) are shown.

Figure 5. Bias (ppbv) between model-calculated and aircraft-observed CO for five different pressure layers. Model-calculated CO was interpolated to the location and time of CO measurements from the DC-8 and P-3B. The number of observations taken in each layer is shown in parentheses. Results from experiments Base, Bbb, Boh, Eoh, SGbb, and BnC are identified with the numbers 1-6, respectively. For each layer, the leftmost six values were calculated using *a priori* CO sources, and the rightmost five values were calculated using *a posteriori* Asian sources.

Figure 6. Taylor diagrams constructed using model-calculated [experiments Bbb0 (a), Base0 (b), SGbb0 (c), and BnC0 (d)] and measured CO from local DC-8/P-3B flights. Model performance is shown for P>850 hPa (1), 850 > P > 700 hPa (2), 700 > P > 500 (3), 500 > P > 300 (4), and P < 300 hPa (5). For each layer, the σ of the observations is plotted in bold along the abscissa. In addition, for each layer, the σ of model output is given by the radial distance between the layer number (in normal font) and the origin; the correlation coefficient is given by the azimuthal position of this number; and the RMS_c is given by the absolute distance between the bold and normal font layer numbers.

Figure 7. Mean latitudinal and vertical distributions of CO calculated from DC-8/P-3B measurements and model output at the same location. Thick solid lines show measured CO. Dotted (thin solid) lines show model-calculated CO from experiments with *a priori* (*a posteriori*) CO sources. The contributions of *a posteriori* ff emissions (dashed lines),

bf emissions (dashed triple dotted lines), and bb emissions (dashed-dotted lines) to model-calculated CO are also shown. a) Latitudinal distribution from experiment Bbbr, b) Latitudinal distribution from experiment SGbb, c) Vertical distribution from experiment Bbbr, d) Vertical distribution from experiment SGbb.

Figure 8. Mean model-calculated (experiment Bbbr2) and observed CO (asterisks) along P-3B flight tracks. Shading shows mean contribution of various source terms and/or regions to total model-calculated CO for each flight. Total CO from experiment Bbb0 is shown with plus symbols. Flight numbers (dates) are shown above (below) individual bars. Average contribution of various source terms over all flights is shown across from source type. a) Partitioned by source type; b) Partitioned by source type and/or region (Asian vs. rest-of-the-world).

Figure 9. Taylor diagrams constructed using model-calculated (in bold font) and aircraft-measured (in normal font along the abscissa) CO. a) DC-8 Flights 7-17 Experiment Bbb0, b) DC-8 Flights 7-17 Experiment Bbbr2, c) P-3B Flights 8-19 Experiment Bbb0, and d) P-3B Flights 8-19 Experiment Bbbr2.

Figure 10. Taylor diagrams constructed using model-calculated (in bold font) CO and measurements (in normal font along the abscissa) from DC-8 flights 7-17. a) Experiment Bbb2 and b) Experiment SGbb2

Figure 11. Mean model-calculated and MOPITT-observed CO columns for March 2001. a) MOPITT data after aggregating onto the 2º x 2.5º CTM grid. b) Model-calculated output from experiment Bbb0 after processing with the MOPITT averaging kernel. c) Model-calculated CO from experiment Bbbr2 after processing with the MOPITT averaging kernel. d) Model-calculated CO from experiment Bbbr2 at location of MOPITT observations but before processing with the MOPITT averaging kernel.

Figure 12. Mean model-calculated and MOPITT sampled CO profiles for March 2001. Model-calculated profiles were passed through the MOPITT averaging kernel. Model-calculated profiles from experiment Bbb0 (Bbbr2) are shown with a dashed (solid) line. MOPITT-sampled values are shown with asterisks. Boxes show values $\pm 1\ \sigma$ of the measured means. Means are shown for 10° boxes centered at the specified longitudes and latitudes.

Figure 13. Mean model-calculated CO with an Asian ff/bf source as a function of pressure for March 2001. Solid lines show model-calculated CO at the location of MOPITT profiles after passing through the MOPITT averaging kernel. Dashed lines show model-calculated CO at the same locations before passing through the MOPITT averaging kernel. Dotted lines show March 2001 mean of all model points within specified latitude/longitude ranges.

Figure 14. Same as Figure 13 but for model-calculated CO with a bb source.

Figure 15. Recent estimates of annual ff/bf emissions from China (Tg CO yr^{-1}) [a] and March 2001 bb emissions from Asia (Tg CO mon^{-1}) [b]. Please consult individual references for information on methods used to calculate uncertainty range.

Table 1. Details of various CO simulations that were performed in this study. .

Sim name	Hor Grid	Notes	OH	Deep conv?
Base0	2.0° x 2.5°	**A**	Spivakovsky	**Yes**
Base2	2.0° x 2.5°	**A**	Spivakovsky	**Yes**
Baser2	2.0° x 2.5°	**A**	Spivakovsky	**Yes**
Bbb0	2.0° x 2.5°	**B**	Spivakovsky	**Yes**
Ebb0	2.0° x 2.5°	**B, C**	Spivakovsky	**Yes**
Bbb2	2.0° x 2.5°	**B**	Spivakovsky	**Yes**
Bbbr2	2.0° x 2.5°	**B**	Spivakovsky	**Yes**
Ebbr2	2.0° x 2.5°	**B, C**	Spivakovsky	**Yes**
BnC0	2.0° x 2.5°	**B**	Spivakovsky	**No**
BnC2	2.0° x 2.5°	**B**	Spivakovsky	**No**
Boh0	2.0° x 2.5°	**B**	Martin/Spiv	**Yes**
Boh2	2.0° x 2.5°	**B**	Martin/Spiv	**Yes**
Eoh0	2.0° x 2.5°	**B, C**	Martin/Spiv.	**Yes**
Eoh2	2.0° x 2.5°	**B, C**	Martin/Spiv.	**Yes**
SGbb0	0.5° x 0.5°*	**B**	Spivakovsky	**Yes**
SGbb2	0.5° x 0.5°*	**B**	Spivakovsky	**Yes**
SG0	0.5° x 0.5°*	**A**	Spivakovsky	**Yes**

*0.5° x 0.5° in region of interest
A. *Kiley et al*. [2003] annually averaged ff/bf emissions used globally. *Duncan et al*. [2003a] climatological monthly average bb emissions used globally.
B. *Streets et al*. [2003] monthly average ff/bf emissions used over Asia. *Kiley et al*. ff/bf emissions used for rest of world. *Woo et al*. [2003] daily bb emissions used over Asia. *Duncan et al*. bb emissions used for rest of world.
C. CO production by AVOC oxidation included.

Note: Experiments using *a priori* CO sources are identified by the suffix '0'. Simulations using *a posteriori* global or Asian CO sources are identified by the suffixes '2' and 'r2', respectively. These calculations use the same spatial distribution of sources as the *a priori* runs; however, the magnitudes of global or Asian direct CO sources is adjusted using the method discussed in Section 3.2.

Table 2. Global (top 7 rows) and regional (bottom 3 rows) correction factors for the ff/bf (A) and bb sources (B) needed to minimize differences between model-calculated and measured CO mixing ratios along DC-8/P-3B flight tracks. Values are shown for **C** = 1. The lower and upper bounds show correction factors for a natural hydrocarbon source equal to 120% and 80% of the model-calculated value, respectively. The fraction of variance explained by the models before and after optimization is shown in column 3 where SSE and SST are the error sum of squares and total sum of squares, respectively.

	A(ff/bf)	B(bb)	1-SSE/SST
Base	1.73 ± 0.07	0.69 ± 0.10	0.858 / 0.920
Bbb	1.51 ± 0.05	1.19 ± 0.19	0.869 / 0.922
BnC	1.44 ± 0.05	1.34 ± 0.16	0.863 / 0.913
Boh	1.43 ± 0.05	1.03 ± 0.19	0.885 / 0.923
SGbb	1.63 ± 0.06	0.77 ± 0.17	0.874 / 0.931
Ebb	1.26 ± 0.04	1.07 ± 0.18	0.904 / 0.922
Eoh	1.20 ± 0.04	0.93 ± 0.17	0.913 / 0.923
Baser	2.44 ± 0.14	0.55 ± 0.10	0.858 / 0.923
Bbbr	1.96 ± 0.10	1.10 ± 0.21	0.869 / 0.923
Ebbr	1.50 ± 0.08	0.98 ± 0.18	0.913 / 0.923